Monday, July 27, 1998 PRESOLAR GRAINS AND ISOTOPIC ANOMALIES I 9:00 a.m. Ussher Theatre

Chairs: R. Strebel U. Ott

Drouart A. Robert F.* Gautier D. Dubrulle B.

Origin of Water in the Solar System: Constraints from Deuterium/Hydrogen Ratios

Sandford S. A.* Bernstein M. P. Swindle T. D.

The Trapping of Noble Gases by the Irradiation and Warming of Interstellar Ice Analogs

Cassen P.* Woolum D. S.

Internal Temperatures of the Solar Nebula Inferred from Astronomical Observations of Circumstellar Disks

Masuda A.* Lu Q.

Isotopic Composition of Molybdenum in Iron Meteorites Viewed from Nucleosynthesis

Choi B.-G.* Huss G. R. Wasserburg G. J.

Oxygen, Magnesium, Calcium, and Titanium Isotopes in Asymptotic Giant Branch and Supernova Oxides

De Laeter J. R.* Rosman K. J. R. Ly C.

Meteoritical Barium Abundance from Carbonaceous Chondrites

Mostefaoui S.* Stadermann F. J. El Goresy A.

In Situ Study of Oxygen Isotopes in Graphite from L3 Chondrites: Indications for Oxygen-Isotopic Heterogeneity

Nuth J. A. III*

Comparison of Non-Mass-Dependent, Chemical, Oxygen-Isotopic Fractionation and Reservoir Mixing Models as Applied to Materials in the Solar Nebula

Podosek F. A.* Nichols R. H. Jr. Brannon J. C. Meyer B. S. Ott U. Jennings C. L. Luo N. *Potassium, Stardust, and the Last Supernova*

Amari S.* Zinner E. Clayton D. D. Meyer B. S.

Presolar Grains from Supernovae: The Case for a Type IA SN Source

Clavton D. D.*

Starting the Growth of Carbon SUNOCONs When Oxygen is Greater than Carbon

Ebel D. S. Grossman L.*

Condensation from Cosmic Gas Made of Free Atoms

Meyer B. S.* Luo N.

Stochastic Histories of Dust Grains Bearing Calcium-48 and Titanium-50 Anomalies

^{*} Denotes Speaker

ORIGIN OF WATER IN THE SOLAR SYSTEM: CONSTRAINTS FROM DEUTERIUM/

HYDROGEN RATIOS. A. Drouart¹, F. Robert², D. Gautier³, and B. Dubrulle⁴, ¹DSM/DAPNIA Physique Nucleaire CEA/Saclay, F-91191, Gif-sur-Yvette, France, ² Laboratoire de Minéralogie, Muséum National d'Histoire Naturelle, 61 Rue Buffon, 75005 Paris, France, ³ DESPA Observatoire de Meudon, F-92195 Meudon, France ⁴ CNRS Observatoire de Midi-Pyrenees, 14 avenue Belin, F-31400 Toulouse, France.

An analytical model of an evolutionary protosolar nebula is used to generate temperature-density radial profiles following the procedure elaborated by Dubrulle (1993). The nebula disk is characterized by its initial mass Md, its initial Radius Rd and the coefficient of turbulent diffusion α . These parameters are constrained by theories of formation of the giant planets: (1) the maximum life time of the solar nebula (1.6 10^7 years), (2) the water condensation temperature should be reached at 5.2 A.U. in 3 10^6 years, (3) the surface density at 19 A.U. must be higher than 15 g cm⁻² in 10^4 years and (4) the angular momentum must be transported outwards to Neptune in less than 10^5 years.

The deuterium enrichment in water relative to the protosolar hydrogen is calculated by an equation of diffusion in this turbulent evolutionary nebula. The evolution of the enrichment factor f as a function of time and heliocentric distance is described by an equation looking as:

$$df/dt = \{exchange\} + \{diffusion\}$$
$$f = (D/H)_{H_2O} / (D/H)_{H_2}$$

The exchange contribution is from [2]:

$$df/dt = k(T) [H_2] \{ \alpha(T) - f \}$$

with k(T) the rate constant of the isotopic exchange reaction : $HD + H_2O \longrightarrow HDO + H_2$

and with $\alpha(T)$ the f value under thermodynamical equilibrium at temperature T.

Two situations were considered: (1) Water was formed in the protosolar nebula by oxidation of H_2 and (2) interstellar water was injected as grains (ice or clays) that subsequently evaporated in the protosolar H2. In situation (1), water reached isotopic equilibrium with H2 in the inner solar system i.e. f \sim 1. During the cooling of the nebula, it is shown that f cannot exceed 1.5 (i.e. -750‰ in δ units). Since most objects (including the Earth) of the solar system exhibit much higher δD values, a solar origin for water must be rejected.

On the contrary, if the initial solar system water was enriched in deuterium (situation 2) with a minimum D/H ratio similar to that measured in some smectites found in the matrix of Semarkona (+3600‰; [3]), typical δD values measured in carbonaceous chondrites are reached within less than 10^6 years between 3 and 5 A.U. Examples of the evolution of f with time and heliocentric distances are shown in the Fig. 1. In this Figure, it can be noted that the minimum f value (f=3) observed in LL3 chondrites as well as f=10 in comets are accounted for by this model.

Therefore, water in the solar system would be mainly of interstellar origin and was not synthesized by the thermal oxidation of the protosolar hydrogen. Such deuterated water was subsequently reprocessed in the turbulent nebula. All the possible nebula calculated in the framework of this theory yield this conclusion.

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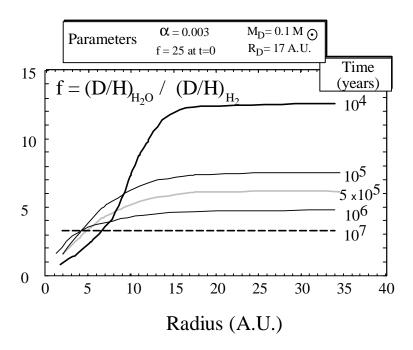


Fig. 1. Isotopic fractionation in water (f) versus the heliocentric distance in the protosolar nebula. Measured f values in solar system objects are 3, 5 (\pm 1), 10 (\pm 2) in LL3 chondrules, mean carbonaceous chondrites and comets, respectively. In this model, water is injected into the solar nebula hydrogen (D/H = 25 x 10⁻⁶) with f = 25.

THE TRAPPING OF NOBLE GASES BY THE IRRADIATION AND WARMING OF INTERSTELLAR ICE ANALOGS. S. A. Sandford¹, M. P. Bernstein¹, and T. D. Swindle², ¹NASA-Ames Research Center, Mail Stop 245-6, Moffett Field CA 94035, USA, ²University of Arizona, Lunar and Planetary Laboratory, Tucson AZ 85721, USA.

Introduction: At the low temperatures typical of interstellar dense molecular clouds (T = 10-25 K), most molecules will freeze out onto grains where they will be exposed to ionizing radiation in the form of UV photons and cosmic rays. The energetic processing of these ices leads to the production of more complex and refractory organic residues [1].

We report recent results in our ongoing effort to determine whether these processes could account for the enigmatic chondritic noble gases found in meteorites [2]. Samples are made at NASA-Ames by simultaneously depositing, at about 10 K, a series of layers consisting of a combination of polycyclic aromatic hydrocarbons (PAHs), CH₃OH, CO, NH₃, and the noble gases He, Ne, Ar, Kr, and Xe. After deposition, each layer is irradiated with UV photons. After the deposition of many layers, the sample is warmed to room temperature and the resulting organic residues are sealed in dry nitrogen and shipped to the University of Arizona for noble gas measurements in a VG5400 mass spectrometer. Originally, the samples were heated overnight to 130-150°C prior to measurement.

Using these techniques, we previously reported very promising results [3], namely concentrations of trapped Kr and Xe that approached within a factor of about 20 of that seen in chondritic meteorites. In this abstract we report work in which we have used isotopically-spiked noble gases to further constrain the gas trapping process.

Results: In the latest set of experiments we have made several changes to our experimental protocol. First, there was some evidence from our earlier work that much of the trapped gas might be escaping during our

130–150°C heatings prior to measurement. In the latest of experiments we have limited the preheating to temperatures below 100°C. In order to better track our sample gases, and to assist with the possible increased contribution from absorbed contamination that might result from our lower temperature prebakes, we replaced our noble gas samples with mixtures that contained isotopically-normal noble gases spiked with additional ²²Ne, ⁴⁰Ar, ⁸⁶Kr, and ¹³⁶Xe.

Conclusions: The results continue to be extremely promising, although there are currently some discrepancies between the trapped gases we produce and those seen in meteorites. On the positive side, we are currently trapping the noble gases Kr and Xe at and above the concentrations seen in meteorites. In addition, the measured Kr and Xe elemental and isotopic fractionations in our samples are very similar to those of meteorites. However, our current samples continue to release the majority of their gases at temperatures below 250°C, a lower temperature than is seen in the meteoritic release. We will be carrying out additional experiments to see whether added photolysis of our samples after warm up makes them more refractory. In addition, the trapping efficiency of Ar relative to Kr and Xe has yet to be established and we are now carrying out several experiments that are optimized to address this issue.

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INTERNAL TEMPERATURES OF THE SOLAR NEBULA INFERRED FROM ASTRONOMICAL

OBSERVATIONS OF CIRCUMSTELLAR DISKS. P. Cassen¹ and D. S. Woolum², ¹NASA Ames Research Center, Moffett Field CA 94035-1000 (cassen@cosmic.arc.nasa.gov), ²Department of Physics, Cal State University, Fullerton CA 92834 (woolum@gps.caltech.edu).

Introduction: If the large scale characteristics of the solar nebula are reflected in the distinguishing characteristics of meteorite classes and components, nebular thermal history is likely to be the primary factor recorded. Until recently, purely theoretical considerations were the main source of information regarding the thermal evolution of the nebula [1]. However, a growing body of astronomical data can now be brought to bear on the issue. The photospheric temperatures of disks around T Tauri stars are constrained by observations at optical, infrared and radio wavelengths [2]. Theoretical models must still be applied in order to use these observations to infer the internal thermal structure of the disks, but the number of stars for which detailed observations and analyses have been performed warrant an assessment of the implications for the thermal evolution of protoplanetary disks like the solar nebula. Complete models should account for obscuring circumstellar matter [3], dust high in the nebular atmosphere [4], and the optically thick nature of the disks themselves. Independent measurements of ultraviolet radiation have been used to estimate disk accretion rates [5,6,7,8], which can be used to further constrain internal disk temperatures. Disks are most likely to be hottest early in their evolution, when they are still heavily obscured by infalling material. In these cases, the total system luminosity provides some estimate of accretional energy, and comparisons with revealed T Tauri luminosities provide a basis for estimating the properties of the embedded disks.

Approach: In order to assess the evidence of revealed T Tauri stars (generally, those with "middle-aged" disks about 1 million years old) regarding internal disk temperatures, we have adopted the following procedure. Estimates of accretion rates obtained from detailed modeling of UV and visible spectra by [6, 7, 8] were used to establish an "accretion temperature" defined in terms of the radiative flux from a steady-state accretion disk [9]. Observed effective temperatures, as well as disk masses, were derived from combined visible, IR and radio observations by [10,11]. We assumed that the observed temperatures represented the combined effects of accretion and externally produced reprocessed radiation of an unspecified nature. The midplane temperature was then calculated from the appropriate sum of radiative

fluxes and the application of simple plane-parallel, gray atmosphere radiative transfer. The optical depth was calculated from the surface density and disk mass, adopting the same model that was used to infer other disk properties from the astronomical observations [10]. Although this procedure involves and approximations, many assumptions application to a set of stars for which the data are internally consistent provides our best determination, on a statistical basis, of the internal thermal state of observed T Tauri disks.

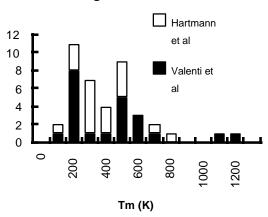
Results: Preliminary results are shown in the Figure, which is a histogram of midplane temperatures at 1 AU for two samples of T Tauri stars. Results are shown separately for accretion rates determined by [6,8]. Most of these disks have internal temperatures of only ~ 200 - 600 K, far less than would be required to vaporize silicate minerals. Midplane vaporization of silicates in these disks would occur at ~ 0.2 - 0.4 AU. However, selection effects tended to exclude stars with the highest accretion luminosities because quantitatively reliable accretion rates are difficult to determine for these stars.

Our approach can be used to estimate observationally consistent internal temperatures for disks at earlier, hotter stages of evolution; at these earlier times, accretion rates have been estimated to be one to two orders of magnitude greater [12,8] during quiescent stages, and even higher during FU Orionis outbursts. It is likely that internal temperatures at distances corresponding to the terrestrial planet region were high enough to vaporize rock-forming elements mainly during these early, higher accretion rate epochs, regardless of whether outbursts occurred or not.

Thus, meteoritic evidence favoring such high temperatures in the terrestrial region of the solar nebula suggests that solid material accumulated rapidly enough into planetesimals to survive from that time. If CAIs are survivors of this epoch and predate chondrules by a few million years, such rapid accumulation could prevent their migration into the Sun, preserving them to be later mixed with the chondrules with which they are now found. Models which produce CAIs and chondrules very close to the Sun [13,14] must preserve precursor solids in their transit to that locale, and so must

invoke much cooler temperatures than those inferred here.





A histogram of midplane temperatures (at 1 AU) of T Tauri stars for which the mass accretion rates, stellar and disk properties have been modelled. Calculations based on data from [6,8,10,11]

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ISOTOPIC COMPOSITION OF MOLYBDENUM IN IRON METEORITES VIEWED FROM

NUCLEOSYNTHESIS. A. Masuda and Qi-Lu, Department of Chemistry, University of Electro-Communications, Chofu, Tokyo 1820021, Japan; correspondence should be addressed to COLGA, 449-1 S-Yasmats, Tokorozawa, Saitama 3590024, Japan (e-mail: masfield@green.ocn.ne.jp)

Molybdenum comprises seven stable isotopes, ⁹²Mo, ⁹⁴Mo, ⁹⁵Mo, ⁹⁶Mo, ⁹⁷Mo, ⁹⁸Mo and ¹⁰⁰Mo, which are different in production process in nucleosynthesis. Isotopic abundance ratios precisely measured in six iron meteorites (Toluca, Gibeon, Canyon Diablo, Hardesty, Landes and Odessa) are dealt with in relation with nucleosynthesis processes; ⁹⁸Mo is taken as a denominator in our mass spectrometric measurement [1]. According to our examination, if ⁹⁵Mo and ⁹⁸Mo are chosen as a normalizing pair and the terrestrial molybdenite values are used as a standard, one can recognize significant relationship between the isotopic relative abundance aberrations. Note that ⁹⁵Mo and ⁹⁸Mo are nuclides produced by s-r(p) and s-r processes [2], respectively. When these two nuclides are taken as a normalizing pair, the following facts are revealed:

(1) There is a positive smooth relation between ε_{94} and ε_{96} with slightly increasing increment with increase of ε_{96} . The relevant curve passes through the earth, which appears to correspond nearly to the average for the iron meteorites studied. Note that 94Mo and 96Mo are produced by p(s) and s processes [2]. This good correlation suggests that the s(p) process may be preferable to p(s) one for the production process of 94 Mo. The value of ε_{94} ranges from 2.81 (Toluca) to -1.80 (Odessa), while that of ε_{96} from 1.94 to -2.33 correspondingly. (2) There appears to be a positive relation between ε_{92} and ε_{96} with drastically increasing increment with increase of ε_{96} ; ⁹²Mo is a p nuclide. The terrestrial point deviates somewhat from the relevant specific curve for iron meteorites. (3) Mostly, ε_{97} is nearly zero; ⁹⁷Mo is s-r nuclide [2] like ⁹⁸Mo and 95Mo which are used for normalization. Four iron meteorites fall in a narrow range (-0.26 ~ +0.09 ε). An aberration of Toluca relative to the average level may be due to 97 Tc. (4) Except Hardesty, ε_{100} shows little variation, but there appears to be a negative correlation with very gentle inclination between ε_{100} and ε_{96} ; 100 Mo is r nuclide [2] in contrast with 96 Mo (s nuclide).

Taken altogether, the foregoing observations indicate that the isotopic composition of Mo in iron meteorites is grossly in reasonable agreement with the theory of nucleosynthesis. with limited peculiarity potentially due to the local overdevelopment or isolation during the processes of production of nuclides. The comparative adequacy of ⁹⁵Mo and ⁹⁸Mo as a normalization pair endorses the relatively farreaching isotopic homogenization of these nuclides. Without the effect of 97Tc, 97Mo (another s-r nuclide) must be subjected to a similar homogenization. A remarkably high correlation between 94Mo and 96Mo covering the difference range of ~4.5 ε in use of 95 Mo and 98 Mo as a normalizing pair implies that the heterogeneity in production rate of s-process nuclide was controlled by a simple and longstanding function, and exempt from the homogenization as compared with the s-r nuclides. At the same time, it can be inferred that the production of s-r nuclides is not just a mere mixing of nuclides produced by perfectly separate s and r processes.

Besides, the isotopic heterogeneity of Mo from sample to sample in bulk iron meteorites gives a significant constraint on the genesis of ion meteorites and on the carriers of Mo isotopes. It is obvious that the bulk iron meteorite retains a primary nature.

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OXYGEN, MAGNESIUM, CALCIUM, AND TITANIUM ISOTOPES IN AGB AND SUPERNOVA OXIDES. B.-G. Choi, G. R. Huss, and G. J. Wasserburg, Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena CA 91125, USA (bchoi@gps.caltech.edu).

We reported O and Mg-Al isotopic compositions of 8 presolar oxide grains from Semarkona and Bishunpur [1]. Here we report O, Mg, Ca and Ti isotopic compositions of additional 5 presolar corundum grains from Semarkona, which include four AGB grains and one supernova grain.

The O isotopic compositions of four corundum (SEAL201, SEAL203, SEAL235, SEAL261) indicate that these grains originated from 1-2 solar mass AGB stars with metallicities within $\pm 50\%$ of solar. SEAL201 has $^{17}\text{O}/^{16}\text{O} = 2.5 \times \text{solar}$ and $^{18}O/^{16}O = 0.28 \times \text{solar}$, which can be explained by Cool Bottom Processing [2]. The grain has a large ²⁶Mg excess of 323±96 ‰, which gives an inferred $(^{26}\text{Al}/^{27}\text{Al})_0$ of $(1.87\pm0.59)\times10^{-3}$. It has ^{50}Ti excess of (1.94±0.72) × solar after correction for ⁵⁰Cr interference, while the other Ti isotopes are normal within errors. SEAL203 is enriched in ^{17}O (7.4 × solar) but has a normal ¹⁸O/¹⁶O ratio. The inferred (²⁶Al/²⁷Al)_o is $(1.03\pm0.56)\times10^{-3}$. The grain has a hint of ^{50}Ti excess with ${}^{50}\text{Ti}/{}^{48}\text{Ti} = (1.24 \pm 0.19) \times \text{solar}$. SEAL235 has $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ ratios of 4.6 and 0.7 × solar, respectively, but has no resolvable ²⁶Mg excess within error (upper limit of $(^{26}Al)^{27}Al)_0 \le 1.0 \times 10^{-4}$). The grain shows ${}^{50}\text{Ti}/{}^{48}\text{Ti}$ ratio of $(2.18\pm0.72) \times \text{solar}$. The Ti isotopic patterns of these three grains are consistent with mixing a small amount of S-process Ti from the He shell (compositions calculated by [3]) with normal Ti from the envelopes of low-mass stars during the TP-AGB stage. The high inferred (²⁶Al/²⁷Al)₀ ratios of the first two grains are consistent with AGB mixing models, however, that of SEAL235 is lower than the models predict. SEAL261 has a high 17 O excess (7.8 × solar) with relative large 18 O depletion $(0.42 \times \text{solar})$. The inferred $(^{26}\text{Al})^{27}\text{Al})_0$ is $(3.9\pm2.8)\times10^{-4}$. In contrast with other grains, it is enriched in ⁴⁸Ti (or depleted in the other Ti isotopes) relative to solar.

The other corundum grain, SEAL122, has an $^{17}\text{O}/^{16}\text{O}$ ratio of 0.00044± 0.00006 (1.2 × solar) and an $^{18}\text{O}/^{16}\text{O}$ ratio of 0.00634±0.00039 (3.2 × solar), making this grain a very strong candidate for a supernova oxide. The grain has normal ²⁵Mg/²⁴Mg ratio, and excess ²⁶Mg, which gives inferred (²⁶Al/²⁷Al)₀ = $(6.9\pm3.6)\times10^{-4}$. The grain has small but significant excess of 50 Ti ((1.38±0.26) × solar), while other Ti isotopic ratios are normal within $\pm 200\%$. There is no signature of 44Ti, which is produced in explosive nucleosynthesis, as inferred from the normal 44Ca/40Ca ratio. The ⁴²Ca/⁴⁰Ca ratio is also normal. We carried out a mixing calculation for 15 solar mass supernova using data given by [4]. The absence of anomalies in ⁴²Ca/⁴⁰Ca and ^{46,47,49}Ti/⁴⁸Ti ratios and the absence of ⁴⁴Ti indicate that the grain has little input from C/O zone and zones interior to it. Large ¹⁸O excess implies that the grain includes materials from He/C zone. However, the high 18O/16O ratio in the He/C zone ($^{18}O/^{16}O \sim 10^3 \times \text{solar for } 15 \text{ solar mass super-}$ nova) and a C/O ratio >1 [4], require significant dilution by an oxygen-rich zone with much lower ¹⁸O/¹⁶O. The measured isotopic compositions of SEAL122 are generally consistent with mixing ~0.3% of He/C zone materials into the H-rich envelope. Calculated results give C/O ratio ~ 0.4 , $^{18}O/^{16}O$ ratio is $\sim 3 \times$ solar and 26 Al/ 27 Al ratio of $\sim 7.7 \times 10^{-4}$. The model predicts only very small Ca and Ti anomalies (≤ 15 ‰). The origin of the small ⁵⁰Ti excess is, however, not clear.

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METEORITICAL BARIUM ABUNDANCE FROM CARBONACEOUS CHONDRITES. J. R. De Laeter, K. J. R. Rosman, and C. Ly, Department of Applied Physics, Curtin University of Technology, Kent Street, Bentley, 6102, Western Australia, Australia.

The slow (s)-process of neutron capture nucleosynthesis of the heavy elements plays an important role in nuclear astrophysics since its behaviour can be modelled to provide information on the helium-burning phase of red giant stars. Because most of the isotopes on the s-process path are stable, their relevant nuclear parameters can be studied experimentally. These parameters are the absolute isotope abundances, their solar system abundances, and the corresponding neutron capture cross-sections at appropriate astrophysical energies. On the basis of this data, s-process nucleosynthesis offers an important test for models describing the late stages of stellar evolution, supposed to be the s-process site.

The classical model of s-process nucleosynthesis, first described in 1957 [1], is a phenomenological approach which has enjoyed success in deriving information on the physical conditions pertaining to the sprocess [2]. The basis of the classical model is that seed nuclei, comprising the iron peak of nuclides, are exposed to an exponential distribution of neutron fluxes, low enough in magnitude that the beta decay rates control the reaction networks. One of the most important features of the s-process model is that a plot of the product of the s-only abundances and the capture cross-sections as a function of mass number, gives a smooth distribution, apart from mass regions where magic neutron numbers occur [3,4]. As more accurate elemental abundance and cross-section data has become available, the classical model has been tested under increasingly stringent conditions. It is now possible to measure this data at the 3% uncertainty level, and deficiencies in the classical model are now emerging.

New capture cross-section values for Sn and Ba has raised questions as to the validity of the presently accepted solar system abundances of these elements [5,6]. In the case of Sn, the s-process model indicates that the abundance of the s-only nuclide ¹¹⁶Sn is too high by approximately 26%, and in the case of Ba, ¹³⁶Ba is too low by approximately 20%. The meteoritical abundance of Sn in the carbonaceous chondrites Orgueil and Ivuna has been redetermined in two laboratories [7,8] The agreement in the two data sets is excellent without necessitating any significent change to the presently accepted abundance value [9].

On the other hand, the solar system abundance of Ba is not particularly well known, so that a redetermination of the accepted value of 2.34 ppm is essential. The stable isotope dilution mass spectrometric technique has been used to measure the meteoritical abundance of Ba in Orgueil and Ivuna. The results for Orgueil (2.15 ppm) and Ivuna (2.33 ppm) confirm the presently accepted value [9], and implies that better sprocess models must be developed.

Recent measurements of the Nd cross-sections [10] have shown that ¹⁴²Nd is overproduced by approximately 12% by the classical model, normalized to the nearby s-only isotope ¹⁵⁰Sm, compared to the solar abundance. Since the relative abundance ratio of Sm to Nd is known to 1.8% [9], one is forced to the conclusion that the simple assumptions of the classical model are inadequate for describing stellar conditions, and that stellar models for the helium burning phase must be investigated.

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IN-SITU STUDY OF OXYGEN ISOTOPES IN GRAPHITE FROM L3 CHONDRITES: INDICATIONS FOR OXYGEN ISOTOPIC HETEROGENEITY. S. Mostefaoui¹, F. J. Stadermann², and A. El Goresy¹. ¹Max-Planck-Institut für Kernphysik, Postfach 103980, 69029 Heidelberg, Germany (smail@pluto.mpi-hd.mpg.de), ²McDonnell Center for the Space Sciences and Physics Department, Washington University, St. Louis MO 63130, USA.

In a previous *in-situ* study of graphite-bearing assemblages in ordinary chondrites we found large H and N isotopic anomalies in a fine-grained graphite morphology from various chondrites. We interpreted these as the result of ion-molecule reactions in the interstellar molecular cloud (IMC) from which the solar system formed [1,2]. Here we report *in-situ* measurements of O isotopes in graphite from the Khohar and Mezö-Madaras (L3) chondrites.

Oxygen isotopic measurements were conducted with the Washington University ion microprobe (a modified Cameca IMS-3F) using a Cs+ beam at high mass resolving power. Laboratory graphite was used as a standard to determine the instrumental mass fractionation. The meteoritic graphites are finegrained and mixed with Fe-Ni metal (see [2]), and in most cases had been extensively sputtered. Thus, analyses contain contamination from the surrounding phases and the isotopic results must be considered with caution. Here, we report 22 measurements on 6 graphite assemblages from Khohar and 5 from Mezö-Madaras (both L3s).

The results show that all the graphites have normal O isotopic signatures (i.e., no extreme deviation from the terrestrial standard). Since the SMOW-normalized O isotopic composition of the terrestrial standard is not known, there is some uncertainty concerning the absolute δ-values for these measurements. However, the results clearly indicate variations in the isotopic compositions of the different graphites relative to each other in Khohar. These variations (along the terrestrial fractionation line) are on the order of 20‰ for the ¹⁸O/¹⁶O ratio. The graphites from Mezö-Madaras on the other hand have more homogeneous O isotopic compositions.

The isotopic ratios in Khohar vary not only from one assemblage to another, but also within the same graphite assemblage. In one measurement of only ¹⁶O

and 18 O, we found 16 O excesses of almost 100‰ in the first two blocks of a 15 block analysis, with δ^{18} O increasing with time toward a normal ratio. This result hints at the presence of a small phase of isotopically anomalous material, that sputtered away during the measurement. However, since only two isotopes of O were measured, we cannot be certain that the isotopic composition in fact lies off of the terrestrial mass fractionation line. Attempts to reproduce this result with a three-isotope O measurement at the same location were not successful.

Although the data are rather limited, the results reported here are the first in-situ measurements of O isotopic ratios in graphite from chondrites. Whereas H and N in graphite are enriched in D and ¹⁵N [2], O isotopic ratios are normal. This is not unexpected, since O isotopic fractionation by ion-molecule reactions in the IMC is predicted to be small [3].

However, the O isotopic ratios in the graphite are more variable than in bulk ordinary chondrites [4]. This seems to reflect an intrinsic isotopic variation, which may be due to the presence of isotopically different graphite components that did not equilibrate with one another. The possible enhancement in ¹⁶O observed in one measurement of Khohar is intriguing and suggests the presence of an isotopically anomalous phase in the graphite. Unfortunately, because this phase appears to be very small, it cannot be measured in detail with current ion microprobes. Future work will include the measurement of H isotopes in graphite from Mezö-Madaras.

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COMPARISON OF NON-MASS-DEPENDENT, CHEMICAL, OXYGEN ISOTOPIC FRACTIONATION AND RESERVOIR MIXING MODELS AS APPLIED TO MATERIALS IN THE SOLAR NEBULA. J. A. Nuth III, Astrochemistry Branch, Code 691, NASA Goddard Space Flight Center, Greenbelt MD 20771, USA (nuth@gsfc.nasa.gov).

Introduction: Ever since the discovery [1] of widespread, non-mass-dependent, oxygen isotopic fractionation in meteoritic materials the preferred explaination for the observations has been the Reservoir Mixing model in which a ¹⁶O-poor presolar gas and dust reservoir mixes with a ¹⁶O-rich reservoir of grains formed in a nearby supernova. Thiemens [2] had suggested an alternative explanation for these observations based on quantum mechanical grounds but was not able to segregate the products of these enhanced reactions into meteoritic components that would survive further processing in the solar nebula [3]. An excellent summary of the current state of our understanding is the recent review by Clayton [4].

More recently Nuth demonstrated a chemical reaction mechanism that could serve to segregate the products of quantum mechanically enhanced reactions into solids [5]. In this mechanism a refractory grain evaporates as a metal monoxide {e.g. SiO or AlO}, reacts with an oxygen source to form a quantum-chemically stabilized metal dioxide {SiO2 or AlO2} whose vapor pressure is many orders of magnitude less than that of the monoxide. When the metal dioxide condenses it will non-mass-dependently sequester heavy oxygen into the solid and leave the gas enriched in ¹⁶O. Repeated cycles of high temperature vaporization, chemical reaction and condensation will monotonically increase the ¹⁷O and ¹⁸O content of the solid and the ¹⁶O content of the gas.

Contrasting the Consequences: Nuth [5] noted a number of logical consequences of the chemical fractionation mechanism: namely, comets should be ¹⁶O-rich compared to SMOW and more highly processed material should be isotopically heavy (so chondrule and CAI cores should be ¹⁶O-rich compared to their rims) to name just two. Clayton [4] has noted that there are no systematic correlations in the O isotopic data that can be predicted by the Reservoir Mixing model, although after 25 years of measurements there have been a large number of individual reservoirs

postulated to explain the observations. Might these many postulated reservoirs contain information that can be used to decide between the Reservoir Mixing and the Non-Mass-Dependent Chemical Fractionation models? Could both processes play a role in the overall chemical evolution of the solar nebula?

This talk will focus on additional consequences of the chemical fractionation mechanism and on expected correlations between the isotopic composition of a parent body and its chemical composition or processing history. As an example, if the initial water/rock ratio of all Ordinary Chondrites was identical, then the refractory component of LL chondrites was more highly processed than that of L chondrites which, in turn was more highly processed than H chondrites. Alternatively H chondrite parent bodies may have contained more volatiles or unprocessed dust than did those of the L's and these in turn either were wetter or contained more presolar grains than the parent-bodies of the LL chondrites.

Oxygen isotopic composition may depend both on the orbital position of the parent body within the nebula {bodies farther out capture more comets and become ¹⁶O-rich} and on the ability of the parent to retain these volatiles until they can equilibrate {Mars retained a smaller proportion of cometary volatiles than did the Earth, but more than was retained by the asteroids}. An extention of this reasoning is the prediction that Venus should contain more ¹⁷O & ¹⁸O than the Earth.

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POTASSIUM, STARDUST, AND THE LAST SUPERNOVA. F. A. Podosek¹, R. H. Nichols, Jr.¹, J. C. Brannon¹, B. S. Meyer², U. Ott³, C. L. Jennings¹, and N. Luo², ¹Department of Earth and Planetary Sciences, Washington University, St. Louis MO 63130, USA (fap@levee.wustl.edu), ²Department of Physics and Astronomy, Clemson University, Clemson SC 29634, USA, ³ Max-Planck-Institut für Chemie, D-55128 Mainz, Germany.

Variations in the isotopic composition of K, generated by mass-dependent fractionation, by radioactive decay or by nuclear spallation, are not common but not unknown [1–5]. As far as we know, however, there are no published reports of *isotopic anomalies* in K, *i.e.* compositional variations reflecting incomplete homogenization of different nucleosynthetic components. But the literature relevant to possible isotopic anomalies is sparse [1,6,7] and because the absolute abundance of ⁴⁰K is so low (0.01%), available data for its relative abundance are not very precise (one half to one percent).

We report here thermal ionization isotopic data with analytical error limits on discriminationcorrected 40K abundances about an order of magnitude smaller than those in prior literature. We have analyzed K in samples generated by progressive leaching of carbonaceous chondrites (Orgueil and Murchison), samples previously found to exhibit pervasive anomalies in Cr [8-9]. Most samples have K compositions normal within error limits, but some, those from HNO₃ or first HCl leaches, appear to have well resolved excesses of ⁴⁰K, up to about 35 ε-units, which evidently require interpretation as isotopic anomalies. As for Cr this most likely reflects the presence of presolar K-bearing grains with various nucleosynthetic histories. The K and Cr anomalies do not correlate in detail and apparently reflect different grain carriers.

The K anomalies contribute little to whole-rock inventories, however; *whole rock* ⁴⁰K abundances in Orgueil and Murchison are normal within 1 permil. This is a non-trivial result because it sets useful limits on the large-scale distribution of late nucleosynthetic input of K to the solar nebula.

The presence of short-lived radionuclides such as ²⁶Al demands input of fresh nucleosynthetic products,

arguably from a supernova [10], shortly (within about 1 Ma) before, and perhaps inducing, molecular cloud collapse. It is frequently hypothesized that ²⁶Al and other short-lived radionuclides may have been very heterogeneously distributed in the nebula, perhaps because of such late input [11]. Potassium-40 provides a usefully sensitive test of the distribution of late nucleosynthetic input simply because it is radioactive, whence most (a detailed model suggests 62%) of the ⁴⁰K ever added to the sun's predecessor material would already have decayed by the time of solar system formation. If a specific 25_⊙ supernova model [12] is invoked to account for 26Al, it should also account for about 1% of stable K and 4% of ⁴⁰K. A planetary body which includes this last input would thus be about 30% anomalous relative to one which does not. Orgueil, Murchison and the earth differ by no more than 1 permil in ⁴⁰K and so cannot have very different amounts of such a late supernova component. Unless an efficient global chemical separation mechanism is postulated, the same conclusion applies to ²⁶Al. This approach should also be applicable to other models for synthesis of interesting radionuclides co-produced with K.

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PRESOLAR GRAINS FROM SUPERNOVAE: THE CASE FOR A TYPE Ia SN SOURCE. S.

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Low-density graphite, Si₃N₄ and SiC grains of type X have common isotopic signatures, mostly isotopically light C, heavy N, and high ²⁶Al/²⁷Al ratios of up to 0.6 (for a summary see [1]). Large ²⁸Si excesses and evidence for ⁴⁴Ti strongly argue for a SN origin. Type II supernovae have previously been proposed as their stellar sources. Travaglio et al. [2] and Zinner et al. [3] mixed different zones of such stars in an attempt to reproduce the grain data. A fundamental problem of the Type II hypothesis is that, if one imposes the constraint C/O>1 to condense carbonaceous grains, it requires mixing of material in the outer zones, which have C/O>1, with that of the innermost Si-rich zones where ²⁸Si and ⁴⁴Ti are produced, these zones being separated by a huge layer that mainly consists of O. Recently, Clayton et al. [4] have suggested Type Ia supernovae as a stellar source for SiC X grains. In their model, essentially all isotopic features exhibited by the SN grains are produced by a single nucleosynthetic process, explosive He burning. Here we explore this possibility in more detail.

In the SNIa model, material having experienced CNO burning accretes from a companion star onto a CO white dwarf where this He-rich cap ignites and burns explosively. Two reaction paths are required to explain the grain data: 1) Successive αcapture from ¹²C produces ²⁸Si and ⁴⁴Ti. 2) Reactions on ^{14}N produce ^{15}N from (p,γ) [in addition to $^{18}O(p,\alpha)$], and ^{18}O and ^{26}Al by α -capture. This requires the presence of ¹⁴N at the time of ignition. Isotopic yields are quite sensitive to peak temperatures and mixing of explosive components of various temperatures is required to reproduce the grain signatures. Even then, the isotopic ratios produced by explosive nucleosynthesis, in particular the C, N and O ratios, are too extreme. We therefore mixed the explosive products with material before He-burning to see whether a satisfactory match can be produced.

The results can be summarized as follows. The most satisfactory match is achieved for the Si isotopes and the $^{44}\text{Ti}/^{48}\text{Ti}$ ratio. For Si the trend of the data is obtained only for $\leq 1.5 \, \text{T}_9$ and high enough $^{44}\text{Ti}/^{48}\text{Ti}$ ratios for $\geq 1.2 \, \text{T}_9$. However, an appropriate mix of contributions from burning at $0.5-1.4 \, \text{T}_9$

can reproduce most of the experimental ranges for C>O, except for the highest 44Ti/48Ti ratios. Even then, ²⁹Si/³⁰Si ratios in the grains are larger than in the SN mix, the same problem as that faced with Type II models [2,3] Mixes of burning products at 0.7–1.0T₉ with original He-cap material (which already contains 26 Al at the level of $0.4 \times ^{27}$ Al) can reasonably account for many observed ²⁶Al/²⁷Al ratios except for low values and for the highest ratio achieved in the grain with the lowest ¹⁴N/¹⁵N ratio. The most serious problems are encountered for the ¹²C/¹³C ratios in combination with ¹⁴N/¹⁵N and ¹⁶O/¹⁸O ratios: the condition C>O is satisfied only for 12C/13C>600, while most grains have smaller ratios, and a reasonable match to the data can only be achieved for C>O. Recently, Clayton [5] proposed that in a SN environment the CO molecule is not stable and that carbonaceous grains could condense from a gas with O>C. It remains to be seen whether this is possible and, if so, both Type Ia and Type II SN models have to be reevaluated.

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STARTING THE GROWTH OF CARBON SUNOCONS WHEN OXYGEN IS GREATER THAN

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In an expansion of gases maintaining thermal and chemical equilibrium, C dust cannot condense unless the C abundance exceeds that of O. During cooling the CO molecule becomes stable before dust is stable, resulting in the early total conversion of C to CO, which is almost inert. The limitation C>O does not apply within supernovae, either SN Ia or SN II. The supernova gas does maintain a thermal ion temperature but cannot achieve equilibrium with its because radioactivity disrupts molecules. The well-studied case of CO molecule [1,2] shows that this disruption may be through either direct reactions with Compton electrons or with the degraded gamma rays, or it may involve chemically disruptive reactions with highly energized species such as He⁺ [1] that owe their existence to the radioactivity. Other diatomic molecules are likewise destroyed, raising the question of how solids grow if diatomic molecules are rapidly destroyed.

Once started, C solids continue growth if the kinetic reactions of oxidation (destructive) are slower than the kinetic reactions of C-cluster growth [2]. In that case, the survival of C solids when O>C may then be understood, because the steady bath of free C atoms maintains growth. This abstract addresses how the growth is initiated in the absence of the thermal phase transition to diatomic molecules as the kinetic temperature falls. The answer is that although the total conversion to molecules is prohibited by the ⁵⁶Co, there exists a steady-state radioactive concentration (near 1%) of diatomic molecules given by the approximate balance between formation and destruction of the molecule. At a characteristic time during the expansion, the lifetime of the CO molecule, for example, is only about $t_{co} = 10$ days, whereas the lifetime of the C atom against conversion to CO is about $t_c = 200$ d. As a result, the steady concentration of CO is but a few percent of the steady concentration of C atoms, and the final CO mass 50 times less [1] than what would have been expected without the radioactive excitations of the system. As the gas cools, the very abundant C atoms establish a supersaturated C atmosphere, the temperature during this steady-state balance falling well below that at which the conversion of C to CO would normally be complete. This is not a thermal phenomenon but a kinetic one.

The same physics applies to other molecules. The radiative association C+C= C_2 can progess with comparable speed owing to the cool (<2000K) atomic-carbon vapor. That the C_2 dissociation energy D=6.25

eV is less than for CO (D=11.09 eV) does not have the relevance for cooled supernova interiors that it would have in a thermal equilibrium, so that a ratio $n(C)/n(C_2) = 50$ or so may be expected within the supersaturated C vapor for this molecule as well. This large abundance of C₂ is the key to the growth of C solids. Radiative association can produce C₃, C₄ and longer chains in sequence, establishing a small abundance of those molecules[3]. It must be investigated if C₂ can react with itself to make C₄ directly, since the C2 abundance (2% of C) is quite high. These small C chains are destroyed both by oxidation and by the Compton electrons; however that only reduces the steady concentration of each. Those steady concentrations seed the growth toward macroscopic weights. The growth accelerates when the cluster is large enough that a C atom can simply stick without need of a process for immediately carrying off the excess energy. Large clusters (nanograins) have sufficient heat capacity to stick a C atom and later radiate (as infrared) the excess excitation. Following this transition, solids become more stable than molecules, even than CO. Although this description is superficially similar to classical nucleation theory, it is really very different owing to the cool supersaturated atomic-C vapor and the very large abundance of C molecules, allowing a fast cascade toward larger sizes in a small fraction of the C mass. Only a small fraction of SUNOCON dust is needed, because the meteoritic evidence requires only a ppm or so of C to condense within SUNOCONs [4].

These same principles, when mirrored, also enable supernova oxides to grow within gas having C>O. Evidence of such a SUNOCON appeared at LPSC98 in the form of corundum grain having large excess ¹⁸O [5]. This large ¹⁸O excess exists in the He-burning shell, which is also the only supernova shell having C>O [6], a composition that would otherwise be expected to prevent corundum condensation. It may be concluded that within supernovae the relative abundance of C and O does not determine whether carbon or oxide grains may condense, as it does in thermal equilibrium.

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CONDENSATION FROM COSMIC GAS MADE OF FREE ATOMS. D. S. Ebel¹ and L. Grossman^{1,2}, ¹Department of the Geophysical Sciences, 5734 South Ellis Avenue, University of Chicago, Chicago IL 60637, USA (debel@midway.uchicago.edu), ²Enrico Fermi Institute, University of Chicago, Chicago IL 60637, USA.

Introduction: It has long been known that the mineralogy of condensates from cosmic gases is strongly dependent on the atomic C/O ratio because of the great stability of the CO molecule [1]. If C/O>1 as in C-rich stars, all O is combined as CO and the excess C is available to form graphite and carbides as high-temperature condensates. If C/O<1 as in O-rich stars, all C is combined as CO and the excess O is available to form oxides and silicates as hightemperature condensates. In meteorites, a significant fraction of interstellar graphite, and SiC grains of the X-type, are thought to have condensed from Type II supernova ejecta whose C/O ratio >1. In order to explain details of the isotopic compositions of these grains, matter from interior zones of the supernova must be mixed with matter from the exterior. Mixing calculations which quantitatively reproduce the isotope ratios, however, invariably give C/O<1 for the ejecta, from which graphite and SiC cannot condense [2]. As a way out of this predicament, Clayton [3] has suggested that supernova ejecta are so intensely irradiated that CO and other gas phase molecules are nearly completely dissociated, liberating C atoms from CO and making them available to form graphite and carbides, even in O-rich ejecta. While it is difficult to envision irradiation conditions which would prevent two atoms from bonding together to form a molecule while still allowing thousands of atoms to bond to one another to form a grain, we nevertheless performed condensation calculations from a gas of solar composition (C/O = 0.42) to see what condensate phases would form at equilibrium under two sets of conditions: (1) CO is prevented from forming; and (2) all polyatomic molecules are prevented from forming in the gas.

Technique: Calculations were done for a fixed total pressure of 10^{-6} bar using the method of [4]. Runs were performed without the silicate liquid models described in [4], and were terminated when refractory elements such as Ti became depleted in the gas phase to a mole fraction of $<10^{-20}$.

Results and Discussion: When the CO molecule is prevented from forming, most of the O forms CO_2 instead, reducing the partial pressure of O by slightly more than one log unit at 1570 K. Consequently, the phases corundum, hibonite, perovskite, melilite, spinel, pyroxene and olivine all appear at temperatures 40° to 60° lower than in a solar gas with CO

present. Simple removal of CO does not cause graphite or carbides to become stable in a gas of solar composition.

When all molecular species are prevented from forming, strange things happen. As predicted by Clayton [3], the C liberated from CO does condense as graphite, at 2110 K. The O liberated from CO, however, is available to form oxide and silicate condensates at higher temperatures than in a fully speciated solar gas, in which the SiO molecule stabilizes Si in the vapor phase. In the present case, the free Si combines with the free O to condense cristobalite (SiO₂) at 1860 K, a higher temperature than that for any other refractory oxide, and after 99.6% of the C has already condensed as graphite. At 1820, Ti₄O₇ condenses and is replaced by TiO₂ at 1790K, where mullite (Al₆Si₂O₁₃) also appears. Anorthite (CaAl₂Si₂O₈) appears at 1770 K, and mullite disappears at 1750 K. Sphene (CaTiSiO₅) replaces TiO₂ at 1700 K, wollastonite (CaSiO₃) appears at 1680 K, and whitlockite (Ca₃(PO₄)₂) at 1610 K. Wollastonite is replaced by diopsidic pyroxene (CaMgSi₂O₆) at 1590 K. Cristobalite disappears at 1530 K, just before forsteritic olivine (Mg₂SiO₄) finally appears at 1520 K. In a solar gas composed entirely of monatomic species, at 10⁻⁶ bar total pressure, graphite and cristobalite are the most abundant condensates between 2100 K and

Suppression of the polyatomic molecular species in a solar gas at 10⁻⁶ bar total pressure changes the order of major element condensation from Al-Ca-Ti-Si-Mg to C-Si-Ti-Al-Ca-Mg. The mechanism called upon by Clayton [3] is unlikely to yield SiC or TiC grains, even if grains could nucleate in an environment which destroys gaseous molecules. Our calculations do suggest that, instead of SiC and TiC, SiO₂ would cocondense with graphite. One might expect extrasolar graphite to be accompanied by extrasolar silicon dioxide, if graphite did indeed form in the manner Clayton [3] suggests. Inclusions of SiO₂ have not been discovered in searches of extrasolar graphite.

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STOCHASTIC HISTORIES OF DUST GRAINS BEARING ⁴⁸**CA AND** ⁵⁰**TI ANOMALIES.** B. S. Meyer and N. Luo, *Department of Physics and Astronomy, Clemson University, Clemson SC 29634, USA (brad@cosmo.phys.clemson.edu)*.

Certain calcium-aluminum-rich inclusions (CAIs) in primitive meteorites bear anomalies in ⁴⁸Ca and ⁵⁰Ti. Both positive and negative anomalies are present, and they are correlated in the sense that excesses of ⁵⁰Ti accompany excesses of ⁴⁸Ca and deficits in ⁵⁰Ti accompany deficits of ⁴⁸Ca (see e.g. the review by Lee [1]). Because ⁴⁸Ca and ⁵⁰Ti can be co-produced in low-entropy, neutron-rich expansions of hot matter, the correlated anomalies call for chemical memory of their nucleosynthesis. We are investigating this chemical memory from the original nucleosynthesis through transport in the interstellar medium (ISM) in an effort to catalog the anomaly-bearing dust.

Recent work has clarified the origin of these two isotopes. It is now evident that ⁴⁸Ca cannot be made in the highentropy environments of core-collapse (Type II) supernovae (explosions of massive stars). Rather this isotope probably is only made in certain rare thermonuclear explosions of white dwarf stars (Type Ia supernovae) [2,3,4]. These explosions have the requisite low entropy. Roughly 2/3 of the solar system's ⁵⁰Ti comes from the same events. The remaining 1/3 comes from massive stars. Crucially, this latter ⁵⁰Ti would coexist with abundant ¹⁶O and ²⁶Al (see e.g. [5]).

In our models, we allow dust to condense from both stellar environments. The Type II SN grains are enriched roughly 15-fold or more relative to solar in ⁵⁰Ti, but are a factor of about two underabundant in ⁴⁸Ca (old ⁴⁸Ca only). For the Type Ia grains, we take an appropriately scaled multi-zone-mixing model using the calculations from [6] (see http://photon.phys.clemson.edu/tables.html). The largest enrichments in ⁴⁸Ca and/or ⁵⁰Ti were roughly 10⁶ in these grains. We inject these grains into the ISM and follow their evolution with a stochastic grain evolution code.

Our model is inspired by previous work [7]. In our model the ISM consists of a diffuse phase at high temperature and low density (in which grains are sputtered) and a dense-cloud phase at low temperature and higher density (in which grains condense mantles). Dust grains cycle between these phases, alternately being sputtered and then accreting matter. Sputtering causes a loss of ⁴⁸Ca-and-⁵⁰Ti-enriched matter, while accretion adds ISM-mixed (hence, more normal) material. Thus, with time, individual dust grains in the dense-cloud phase become less anomalous. Furthermore, more dust grains disappear due to sputtering, but a certain lucky few survive with large mantles.

Fig. 1 illustrates the time evolution of the overabundance of ⁴⁸Ca in a particular Type Ia grain. The overabundance of

⁴⁸Ca is the ratio of the mass fraction of ⁴⁸Ca in the grain relative to the solar mass fraction. Initially the overabundance was 10⁵. We assume the ⁴⁸Ca to be uniformly distributed throughout the refractory core; therefore, sputtering of the core does not change the ⁴⁸Ca overabundance. Sputtering of the mantle, on the other hand, removes normal material and increases the overabundance. Accretion, which we take to be nearly instantaneous, depletes the overabundance by adding more normal matter. The interesting result is that this grain retains much of its large ⁴⁸Ca overabundance even after 10⁹ years.

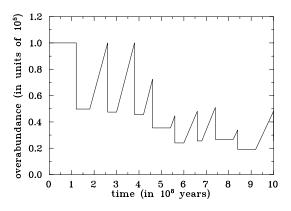


Fig. 1.

We have followed ensembles of individual grain histories for both Type Ia and II SN grains. We have cataloged the results. The results can be compared to the correlated anomalies. Our next task will be coupling our grain evolution and galactic abundance evolution codes to attempt a more complete picture of the history of these grains.

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